PICOSECOND SINGLE ELECTRON PULSES OF 35 MeV TODAI-LINAC AS IRRADIATION SOURCE

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A picosecond single pulse radiolysis system for emission spectroscopy with a response down to 18 picoseconds(ps) was achieved by a combination of fast detection techniques and an intense picosecond single electron pulse produced by 35 MeV S-band Todai-Linac.(1) The technique of picosecond single pulse radiolysis will provide a very strong means of studying on early events in radiation chemistry, radiation biology, and radiation damage of ionic crystals.

During a span of 18 years, early events in radiation-induced reaction have been observed in shorter and shorter times, starting with microsecond techniques, then followed by nanosecond and picosecond techniques. Picosecond pulse radiolysis has been started by using stroboscopic techniques, which detect the absorption of reactive species at times from 20 to 350 ps.(2) Then radiation-induced emission has been detected by a biplanar phototube at times from 60 to 770 ps.(3) The use of a train of picosecond pulses in above-mentioned two cases leads to an uncertainty in age of a radiation-induced primary product, if the product does not decay to zero between the pulses. Recently the absorption of reactive species has been detected at times from 100 ps to 3.5 ns by using single pulse stroboscopic pulse radiolysis, although there is still the long-time limitation of 3.5 ns due to the necessity of using Cerenkov radiation at analyzing light.(4)

We have modified the above-mentioned picosecond pulse radiolysis systems to gain two significant experimental advantages. First, the decay of reactive species can be measured by only one shot of picosecond single electron pulse and the fast detection system, although sampling system is essential for above-mentioned picosecond pulse radiolysis systems. Since a flow system is necessary to avoid overheating and builtup of radiolytic products for sampling system, a lot of fresh solutions are necessary to be circulated through an irradiation cell. Second, since a combination of fast detection system and picosecond single electron pulse is used, there is no long-time limitation for this picosecond single pulse radiolysis.

REQUIREMENT FOR ELECTRON BEAM AND DETECTION SYSTEM

The following characteristies of electron beam are required for this picosecond single pulse radiolysis system.

(1) Single pulse without a satellite micropulse on either side of the main pulse (as shown in Fig. 5)
(2) Intense pulse enough to produce detectable concentration of reactive species in irradiated sample.
(3) Focusing the electron beam to increase the current density.
(4) Stability of the linac. (beam current and beam spot)
(5) Simple operation of the linac.

The following characteristics of the detection system are required.

(1) A very fast response
(2) A wide-time range detection
(3) Small jitter of the dection system itself.

(4) Small timing jitter between the detection system and the picosecond single pulse

PICOSECOND SINGLE PULSE RADIOLYSIS SYSTEM

Fig. 1 shows the schematic diagram of picosecond single pulse radiolysis system for emission spectroscopy. Fig. 2 shows the response of the picosecond single pulse radiolysis system measured by using Cerenkov light induced from electron fine-structure pulses of 10 ps duration (estimated value) produced by a 35 MeV S-band linac. The observed pulse width is 18 ps. This pulse width is formed from two components: the first one is the pulse width of electron beam itself and the second one is the rise and fall times of the detection system. Fig. 3 shows the integrated signal of Cerenkov light pulses induced from one hundred picosecond single electron pulses. The observed pulse width is about 70 ps. This indicates the value of the timing jitter between the detection system and the picosecond single electron pulse in addition to the time resolution of the detection system and the real width of the single electron pulse.

STUDIES ON EXCITED STATES IN IRRADIATED LIQUID HYDROCARBON BY USING PICOSECOND SINGLE PULSE RADIOLYSIS SYSTEM

A lot of work has been carried out on excited states of liquid hydrocarbons by absorption and emission spectroscopy. A number of reviews have been published on excited states of hydrocarbons and energy transfer processes.(5) However, little work has been done on kinetic behavior of excited states of liquid hydrocarbons in the picosecond time range. The following subjects have been carried out.

(1) Formation processes of excited states of irradiated liquid hydrocarbons

(2) Energy transfer mechanism from excited solvent molecules to both solvent molecules and solute molecules

Fig. 4 shows the growth of the fluorescence of solute molecules in the picosecond single pulse radiolysis of irradiated toluene solutions by using picosecond single electron pulse as shown in Fig. 5. The growth of the fluorescence of solute molecules corresponds to the formation of excited solute molecules. As a result of analysis of the experimental data such as the curve of the growth of fluorescence, quencher effects, and temperature effects, two different very fast energy transfer processes are found to exist in irradiated liquid hydrocarbons. The rate constants of the slower energy transfer process are an order of $10^{11} M^{-1} sec^{-1}$ as shown in Fig. 4(a) and (b). This reaction rate is one order faster than that of diffusion-controlled reaction in these liquid hydrocarbon solutions. The rate constants of the faster energy transfer process are more than $10^{13} M^{-1} sec^{-1}$ as shown in Fig. 4(c). This reaction rate is three order faster than that of the diffusion-controlled reaction in these liquid hydrocarbon solutions. These two kinds of very fast energy transfer processes provides new concept of mechanism of energy transfer in irradiated liquid hydrocarbons.(6)

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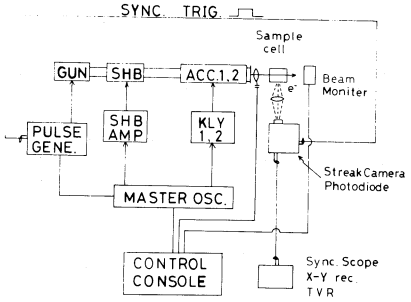

Fig.1 Schematic diagram of a picosecond single pulse radiolysis system

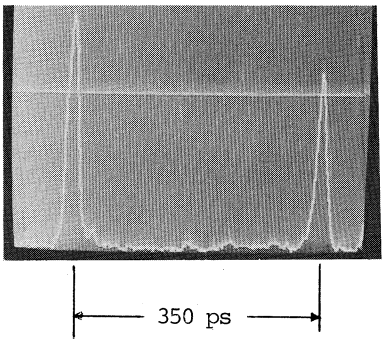

Fig.2 Response of the picosecond single pulse radiolysis system measured by using Čerenkov light induced from electron fine-structure pulses of 10 ps duration (estimated value) Electron fine-structure pulses are produced by a 35 MeV S-band linac. The observed pulse width is 18 ps.

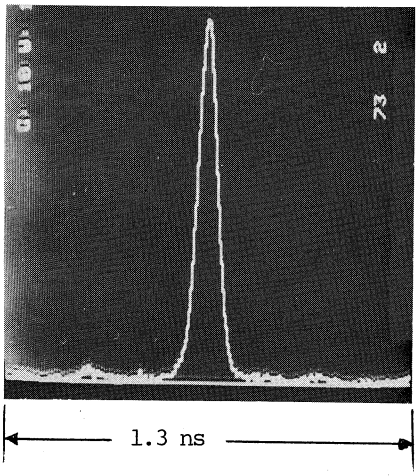

Fig.3 The integrated signal of Čerenkov light pulses induced from electron pulses. One hundred picosecond single electron pulses are used. The observed pulse width is about 70 ps.

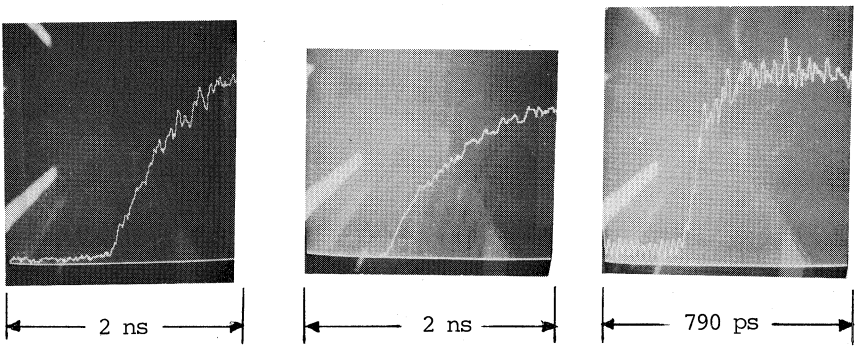

Fig.4 Growth of the fluorescence of solute molecules in irradiated toluene solutions measured by the picosecond single pulse radiolysis system
(a) 10 mM α-NPO toluene solution
the signal obtained from one shot of the electron pulse
(b) 4 mM DM-POPOP toluene solution
the integrated signal obtained from 256 electron pulses
(c) 7.5 mM TPB toluene solution
the signal obtained from one shot of the electron pulse
α-NPO: 2-(1-naphthyl)-5-phenyloxazole
DM-POPOP: 1,4-bis-2-(4-methyl-5-phenyloxazole)benzene
TPB: 1,1,4,4-tetraphenyl-1,3-butadiene

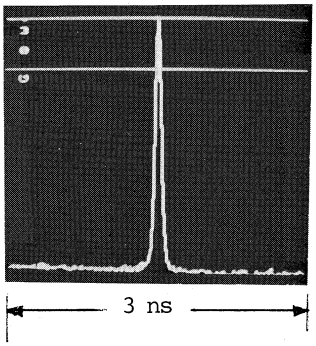

Fig.5 Picosecond single electron pulse achieved by Todai-Linac with subharmonic buncher used for the picosecond single pulse radiolysis system